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6. AUTHOR(S)
Dr Paul H. Cutler

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Department of Physics
Penn State University
104 Davey Laboratory
University Park, PA 16802-6300

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13. ABSTRACT *(Maximum 200 words)*

Theoretical study during this grant period focused on solvable but realistic models of sharp metallic microtips to investigate the significance of curvature on the current-voltage characteristics,, the field emitted electron distributions (FEED) and the thermal stability of these emitters operating in high fields and temperatures. Field electron energy distributions have been calculated for hyperbolidal, paraboloidal and conical models of ultrasharp tips. A calculation was completed for the local density of states (LDOS) of a single atom discrete tip model. The charge simulation method was used to calculate the capacitance of a model diode consisting of a rounded conical tip and an annular ring a distance d above the tip.

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Date: 1 March 1996
From: Paul H. Cutler, Nicholas M. Miskovsky, and T. E. Sullivan
To: Dr. Robert J. Barker
Subject: FINAL TECHNICAL REPORT FOR GRANT NO. AFOSR-91-0408

The attached memorandum reports research progress and accomplishments during the period October 1, 1991 to September 30, 1995 for the proposal, "Experimental and Theoretical Study of High Current Density Novel Emitter Sources," supported by the DoD Vacuum Electronics Initiative and supported by the Air Force Office of Scientific Research under grant AFOSR-91-0408. Theoretical study during this grant period focused on solvable but realistic models of sharp metallic microtips to investigate the significance of curvature on the current-voltage characteristics, the field emitted electron distributions (FEED) and the thermal stability of these emitters operating in high fields and temperatures. In addition, calculations of field emission from diamond were done and confirmed the measured high current emission at low fields but revealed difficulties in identifying viable transport mechanisms to sustain current flow. More specifically, the objectives of this study were (i) to investigate the validity of the Fowler-Nordheim model to describe field emission from the very sharp emitters used in modern microelectronics and technological devices, (ii) to investigate the dependence of FEED on curvature for hyperboloidal, paraboloidal and conical models of ultrasharp tips, (iii) to study heating effects and stability during emission from blunt and ultrasharp tips with particular emphasis on the Nottingham effect and other energy exchange processes, (iv) to study the relationship between tip curvature and thermal stability, and (v) to calculate the capacitance and transconductance of a vacuum field effect transistor. In addition, we also (vi) initiated a detailed study of field emission and transport properties from diamond, including the role of different scattering mechanisms and (vii) calculated the local density of states of a single atom discrete tip model. The latter was done to obtain the necessary input for a fundamental calculation of the current-voltage characteristics of ultrasharp or single atom emitters. This theoretical work was done by the Penn State group.

The experimental group at Temple University (i) worked on fabrication and structural characterization of atomically sharp ($r_t \leq 1$-5 nm) silicon emitters incorporated into MIS point-contact diode configurations, (ii) continued the electrical characterization of the diode and (iii) continued the study of silicide formation on atomically sharp Si tips with the objective of increasing the conductivity of these tips. The group also fabricated metal clad atomically sharp Si emitters using a casting and molding technique. In particular, sharp W emitters were successfully formed using this process with atomically sharp Si tips as precursors. Free standing metal films were produced of these sharp emitters. An improved self-aligned gate process was developed and became the standard process sequence for triode fabrication. Detailed TEM studies of the ultrasharp Si tip structure indicate that the prolate spheroidal tip model replicates the observed tip structure. Hence, an analytic treatment of the electrical and thermal properties developed at Penn State can now be directly applied to analyze experimental data. In addition, a NEXTRAL reactive ion etcher was used for developing an all dry emitter process which significantly improves array uniformity across a "wafer." This work was done at Temple University and at Bellcore Communications Research (Red Bank, New Jersey) where experimental facilities have been made available to students from Temple University working on this project. Since the Bellcore facilities were phased out, these activities took place at Fort Monmouth in New Jersey.

FINAL REPORT ON "EXPERIMENTAL AND THEORETICAL STUDY OF HIGH CURRENT DENSTIY NOVEL EMITTER SOURCES"

P.H. CUTLER, N. M. MISKOVSKY AND T.E. SULLIVAN[a]
THE PENNSYLVANIA STATE UNIVERSITY
UNIVERSITY PARK, PA 16892

Since the development by Spindt of miniaturized vacuum field emitters [1] there has been much experimental evidence to suggest that maximally optimized emitter structures have not as yet been produced. Factors which play important roles in understanding the basic or fundamental mechanisms affecting electron emission are: 1) the detailed surface topology or "shape" of the emitter, 2) the role of thermal effects, 3) materials (including metal silicides, silicon carbide and diamond deposited on atomically sharp silicon) which are potentially important as efficient current sources and 4) the accuracy of the physical and matematical models in predicting the emission current.

Beginning in 1991 we started a combined experimental and theoretical study of new and innovative electron emission sources based upon vacuum field emission and vacuum microelectronic technology. The theoretical part of the study was primarily focused on doing calculations on solvable but realistic models of sharp metallic microtips to investigate (i) the validity of the Fowler-Nordheim model to describe field emission from the very sharp emitters used in modern microelectronics and technological devices, (ii) energy exchange processes and heating during emission from blunt and sharp tips, (iii) the use of field emission energy distributions (FEED) from sharp tips to study the electronic structure of the emitter, and (iv) the transconductance and capacitance (frequency response) in FET vacuum microtriodes. This work was done by the Penn State group.

The experimental group at Temple University did (i) fabrication and structural characterization of atomically sharp ($r_t \leq 1$-5 nm) silicaon emitters incorporated into MIS point-contact diode configurations and (ii) the electrical characterization of the diode. In addition, the group worked on fabrication of metallized emitters using atomically sharp Si tips for a casting and molding technique. In particular, atomically sharp W emitters were successfully formed using this process. Free standing metal films were produced of these sharp emitters. In addition, an improved self-aligned gate process was developed and will become the standard process sequence for triode fabrication. This work was done at Temple University, Bellcore Communications Research (Red Bank, New Jersey) and Fort Monmouth in New Jersey.

[a] Department of Electrical Engineering, Temple University, Philadelphia, PA 19122

A. Theory Group at Penn State University

- The I(V) characteristics for electron emission from metallic emitters with sharp tips ($r_t \leq 10$ nm) were calculated exactly using the kinetic formulation of the tunneling current integral [2]. The I(V) curves are non-linear and show a dramatic increase in the tunneling current for a fixed bias V relative to the Fowler-Nordheim (FN) result (planar geometry) This is illustrated in Fig. 1.

- A β-modified FN equation for the analysis of experimental field emission data is not valid when applied to sharp emitters ($r_t \leq 10$nm). Specifically, it underestimates the emitting area and overestimates the field. These results are illustrated in Fig. 2.

- The resistive heating and the temperature distribution in a prototype (conical) Spindt type emitter have been calculated using a Green function solution for the 3-D heat diffusion equation [4]. The tip heating is a strong function of the cone angle and is localized to the region near the tip apex (see Fig. 3).

- In electron emission, energy exchanges take place between the emitted electrons and the cathode surface. In addition energy transfers between the replacement electrons from the external circuit and the cathode lattice become particularly important at the very high current densities present in field and thermal-field emission. These exchange processes, or the so-called Nottingham effect [5-8] are important in determining the local temperature at the emitter surface. According to Nottingham [5], if the average energy $<\epsilon>$ of the emitted electrons is less that of the replacement electrons, $<\epsilon_r>$, the cathode tends to be heated during emission; if $<\epsilon>$ is greater than $<\epsilon_r>$, the cathode tends to be cooled by the exchange. There exists a temperature, called the inversion temperature, T_i, at which the net heat is zero. Therefore T_i constitutes the equilibrium temperature and helps determine the thermal stability of the emitter. Calculations for sharp tips show that curvature effects can contribute significantly to thermal stability [8]. For example, sharp emitters can operate at higher current density than a blunt tip at the same temperature (see Fig. 4).

- In the Nottingham effect, the energy of the replacement electron is taken to be, under equilibrium conditions, the Fermi energy, ζ_0. This was disputed by Fleming and Henderson [9]; some experimental [7] and theoretical [6,8] results suggest that $<\epsilon_r>$ is less than ζ_0 by at least tens of meV's. We have also developed a more complete and rigorous theory including non-equilibrium effects for the calculation of the average replacement energy of the electrons injected from the external circuit. The results indicate that $<\epsilon_r>$ is lowered by up to 100 meV for fields $\sim 10^8$ V/cm. This resolves a long-standing controversy about the value of the average replacement energy [8,9]. Fig. 5 displays the much improved agreement between measured and calculated values of the inversion temperature, that is, the temperature for stable operation of the emitter.

- Field electron energy distributions (FEED) have been calculated for hyperboloidal, paraboloidal and conical models of ultrasharp tips. These shapes approximate well the

actual or observed shapes of Spindt emitters. The calculated FEED for the curved emitters differs markedly with the FEED curves for the planar FN model. In addition, recent experimental results [10] for FEED from a Molybdenum Spindt cathode is in very good agreement with the calculation for a model emitter for a similar cone angle and temperature emitter which mimics the actual emitter (see Fig. 6). These results indicate that sharp tips allow one to probe energy states much further below the Fermi energy that with "blunt" emitters used in earlier studies. This suggests the usefulness of FEED to elucidate the location and types of tunneling states of new materials proposed for use as field emission sources.

- The conventional treatment of field emission is the Fowler-Nordheim equation which uses a planar model of the emitter. This reduces the problem effectively to one-dimension along the emission direction. It is most convenient theoretically to express the current density J as a function of field F. The predictions based on the relationship, J(F), can only be compared with experimental emitters that are essentially planar (i.e., $r_t > 100$ nm). However, for most current field emission applications in technology, $r_t \leq 100$nm [11]. Some important parameters for the case of sharp tips (for which geometry is important) are the current, applied voltage and beam divergence, which are also experimentally measured quantities. We therefore did model 3-dimensional calculations of the current-voltage characteristics, I(V), for axially-symmetric geometries replicating actually observed tip shapes. In addition to total current calculations, beam divergence and effective emitting areas were also determined. Some characteristic results for r_t=5, 10, and 50 nm and V=20, 50 and 100 Volts are given in Figs. 7, 8 and 9 for a model hyperboloidal W tip with a planar anode. More specifically, beam divergence, dependence of current on tip sharpness and the size of the emitting area as a function of field for different tip radii are given in the figures. These results do not support the conclusions of Spindt et al. [1], that the effective emitting area, as deduced from an analysis using conventional F-N theory based on a planar model tip, is equivalent to a few (~3-5) atomic sites.

- Recent experimental studies of electron field emission from CVD diamond films or diamond coated tips have attracted great attention. This is, in part, due to the fact that large emission currents from diamond can be obtained at low applied fields relative to metals or narrow band-gap semiconductors. Twitchell et al. [12] obtained current densities of 10^{-5} A/cm^2 in the field range from 30-70 V/μm. Zhu et al. [13] recently studied the onset field for field emission from CVD diamond films as a function of defect concentration. Their results indicate a decreasing onset field with increasing defect concentration. For current densities of 10 mA/cm^2, fields of 40-80 V/μm are required. Theoretical understanding of electron emission from diamond surfaces and the related transport processes is still very limited and only now being addressed. In recent papers [14,15] the authors calculated the electron emission using a model consisting of the projection of the energy band surfaces in the emission directions. It is found that the tunneling from bulk conduction and valence bands is negligible in p-type diamond, while copious emission from n-type doped diamond and surface states near the conduction band edge can be obtained. In addition the authors have hypothesized that defect states in the energy gap (shown in

Fig. 10) may be the transport channels of the field emission current. Calculations based on the defect band model yield sufficient transmission probabilities to produce the low power-high current observed in experiments of Twitchell et al.[12] , Zhu et al.[13] and others. The calculated field emission current (depicted in Fig. 11) from defect band states in the gap show a resemblance to the experimental measurements of Twitchell et al. [12] with onset, followed by exponential behavior leading to saturation. Negative electron affinity (NEA) is thought to have a potentially important influence on electron emission from diamond and other wide band-gap semiconductors. Preliminary studies on the role of NEA were begun shortly before the expiration of the grant and are continuing.

- A calculation was completed for the local density of states (LDOS) of a single atom discrete tip model. The tip model is illustrated in Figure 12. In Figure 13, the resulting LDOS is shown. There is at present no fundamental calculation of the tunneling current from an actual discrete tip. Our results will be used as input to obtain for the first time the current-voltage characteristics of ultrasharp or single atom emitters.

- The charge simulation method [16] was used to calculate the capacitance of a model diode consisting of a rounded conical tip and an annular ring a distance d above the tip. The calculated capacitance as a function of geometry is shown in Fig. 14. These results are in agreement with Hill et al. [17] for a similar scaled up device. This method is applicable to multi-electrode devices and is computationally simpler than finite difference methods.

B. Experimental Group at Temple University

- Detailed TEM studies of the ultra sharp Si tip structure indicate that the prolate-spheroidal tip model replicates the observed tip structure. Hence, an analytic treatment of electrical and thermal properties developed at Penn State can now be directly compared to the experimental data (see Fig. 15).

- The following experiment was done to test the theoretical calculations which indicated that field emission currents from atonically sharp field emitters should exceed those predicted by the planar FN equation.

Planar metal-tunnel oxide (<3 nm)-semiconductor (MIS) devices were first fabricated. Atomically sharp emitter tips were fabricated in which only the apex region was exposed with the remainder of the emitter shank enclosed in a thick oxide. The exposed emitter tip was then re-oxidized to grow the same tunnel oxide as the planar devices. The tip tunnel oxide was then metallized and the devices electrically characterized. The exposed emitter tip was examined and measured in a SEM. The current density determined from the current-voltage characteristics and the SEM determined area indicated that J for the atomically sharp tip was nearly 200 times that of the corresponding planar MIS device.

- A NEXTRAL reactive ion etcher is in place and has been used for developing an all dry emitter process which significantly improves array uniformity across a "wafer."

- A revised high temperature process has been achieved through removal of silicon dioxide spin on glass and its replacement with Plasma Enhanced Chemical Vapor Deposited (PECVD) silicon dioxide.

- A planarization and etch back process has been developed for gate formation; a nitride controlled variable gate aperture process has been developed; a newly designed variable array sized photomask is near completion.

- The formation of atomically sharp W tips was achieved using a casting and molding techique. The molds are formed from Si tips with a thermal oxide coating. The process involves the removal of the Si, leaving the impression of the tip in the thermal oxide. Deposition of metal into this mold is carried out and then separation of the cast from the mold yields the metal tip with the same shape as the Si tip used to form the mold. Although these tips are not atomically sharp, preliminary studies indicate that the technique developed can be used to form such tips.

- Using the baseline triode fabrication process developed at Bellcore and currently at Ft. Monmouth ETDL, preliminary studies indicate that the Si emitter tips may be successfully clad with either silicides, carbindes or diamond thin films without sugnificantly blunting the atomically sharp character of the Si emitters. The choice of a Si substrate field emitter technology over metal, diamond, or carbide substrates has two major advantages.

i) Atomically sharpened Si field emitter arrays are formed directly in the Si substrate by stress inhibited oxidation. This use of standard VLSI processing tools such as photolithography, Si etching and Si oxidation ensure that high uniformity may be achieved wafer to wafer and within a wafer.

ii) Once atomically sharp arrays of Si field emitters are formed, materials such as metal silicides, silicon carbind, and diamond may, in principle, be deposited or heteroepitaxially grown on top of the already formed emitter array.

In this way nearly atomically sharp metal, silicon carbide and diamond films can be prepared without the need to attempt the formation of the corresponding sharpened material system. This is particularly advantageous when dealing with hard to etch or shape materials, such as silicon carbind or diamond, materials of technological interest which are well suited for high current and temperature applications, but difficult to form into very sharp emitters.

The emergence of two important and potentially compatible technologied, the fabrication of atomically sharp silicon emitters in conjunction with diamond and SiC film deposition when merged may provide a unique technology capable of addressing the issures of stable, reliable high current and temperature field emission operation at GHz speed.

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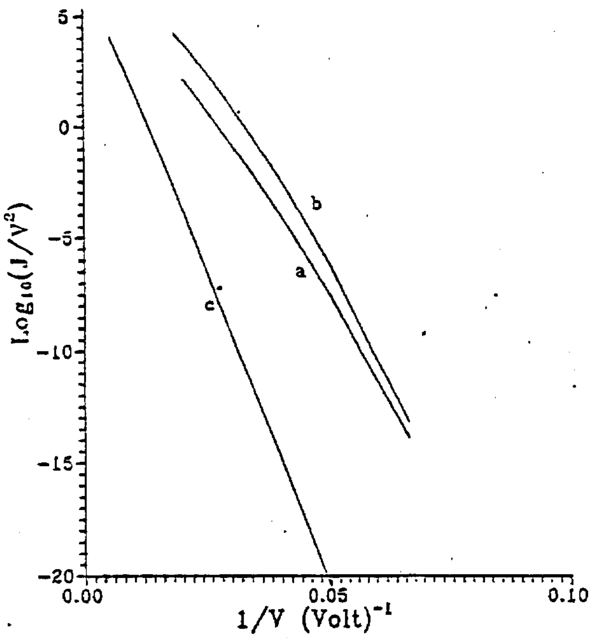

Fig. 1. Plots of $Log(J/V^2)$ vs 1/V for (a) the hyperboloidal model with tip radius of 10 nm, (b) a cone of half angle 70° and (c) the planar model. The tip-collector separation is 20 nm for all models.

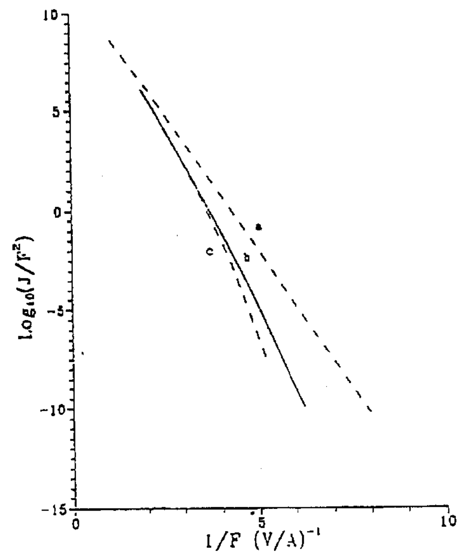

Fig. 2. Plots of $Log(J/V^2)$ vs 1/F for (a) the FN planar model with a β-factor to account for curvature (dashed) and (b) the hyperboloidal model (solid). The radius of curvature is 10 nm. F is the local field at the apex in both models.

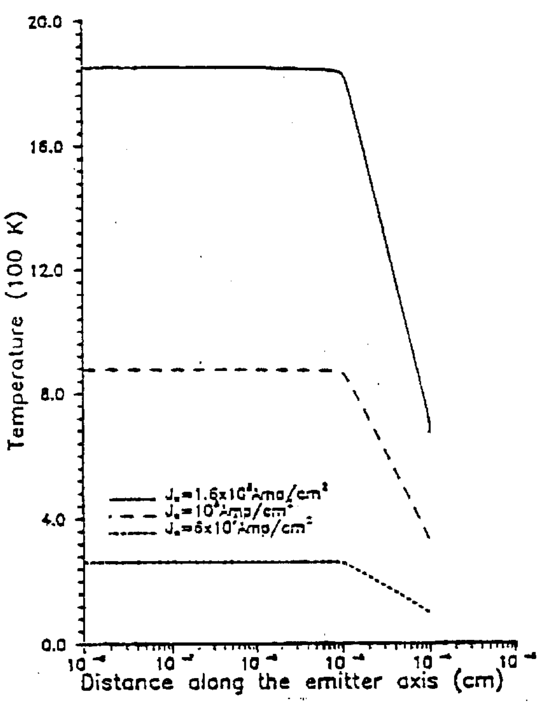

Fig. 3. The temperature rise due to Joule heating vs radial distance along the axis of the emitter for different current densities. The heating time is 100 ns. The resistivity of the W sample was taken to be 5×10^{-5} Ω-cm.

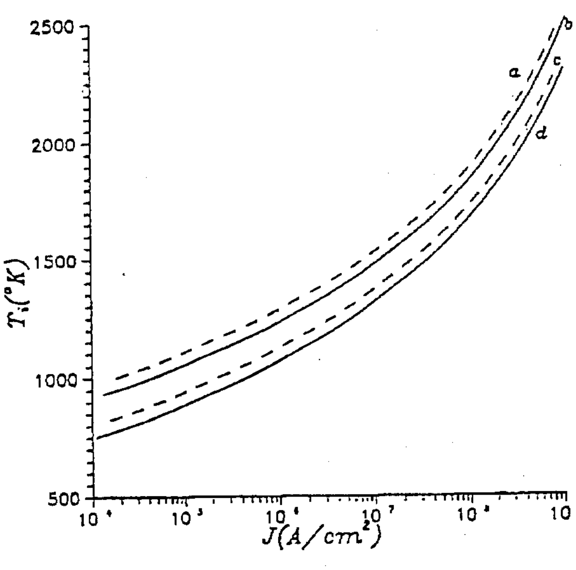

Fig. 4. The Nottingham inversion temperature T_i is plotted as a function of current density J. (a) Planar model with the replacement energy, R=-4.50 eV; (b) hyperboloidal model, R=-4.50 eV; (c) planar model, R=-4.56 eV; (d) hyperboloidal model, R=-4.56 eV.

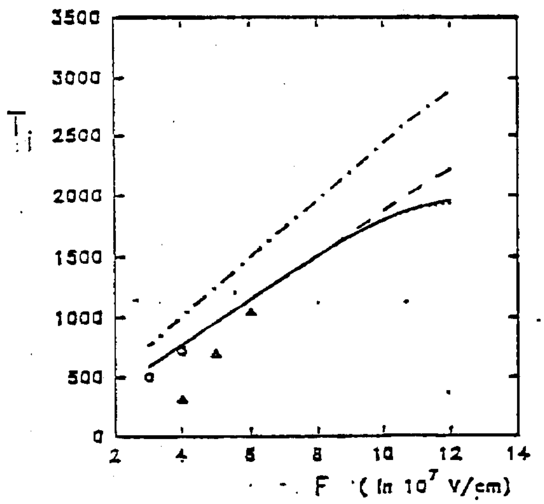

Fig. 5. The inversion temperature T_i as a function of applied field F. The dot-dash curve is the FE theory of Swanson et al.[7]. The dashed curve is calculated using Fleming and Henderson theory [9]. The solid curve is the present theory but including also non-equilibrium effects due to temperature gradients and fields. The experimental data is taken from Ref. 7.

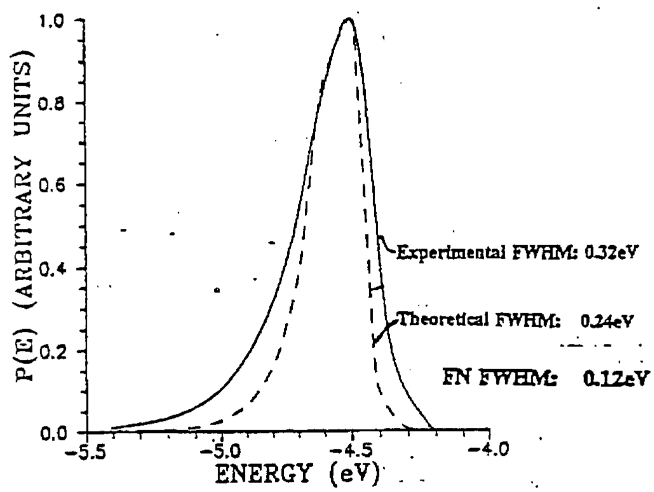

Fig. 6. Total energy distributions (TED) for a W-emitter at 300 K (dashed) and an experimetnal TED for a molybdenum on Si emitter (solid) [10].

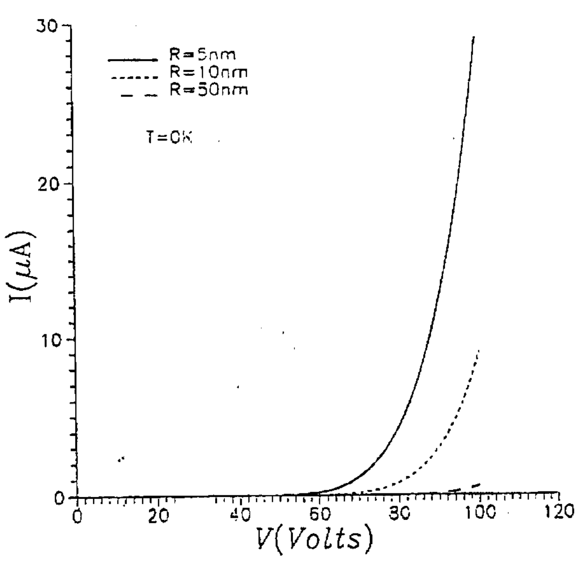

Fig. 7. The I-V characteristics of a three-dimensional hyperboloidal emitter. The tip-collector separation is 20 nm.

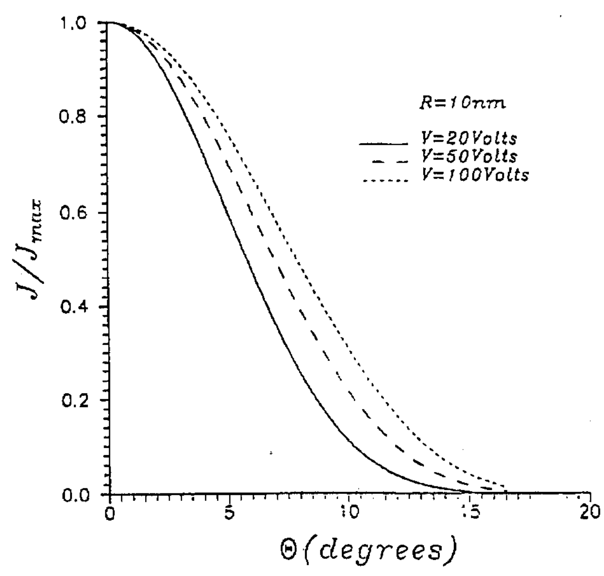

Fig. 8. The ratio of the current density at angle θ from the axis, $J(\theta)$, to the current density on axis J_{max} as a function of θ. The radius of curvature of the tip is 10 nm and the tip-collector separation is 20 nm. For this radius of curvature there is an increase in the effective emitting area as the voltage V increases.

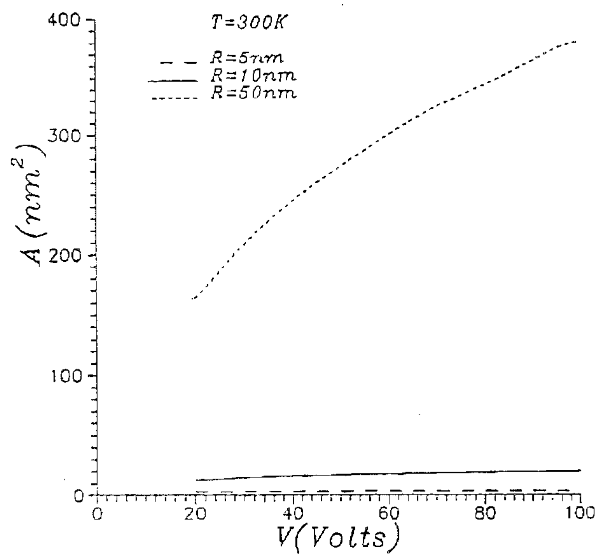

Fig. 9. The effective emitting area A of a hyperboloidal tip (for various radii of curvature of the tip) as a function of applied voltage V.

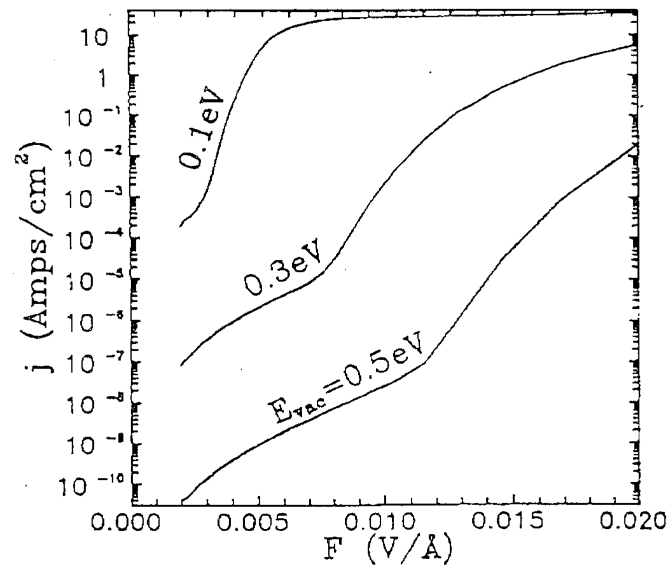

Fig. 11. Plot of j vs F of the field emission for the defect band model. A direct gap of 0.5 eV is assumed. The effective masses are chosen to be the electron mass m_e. The temperature is T=300 K.

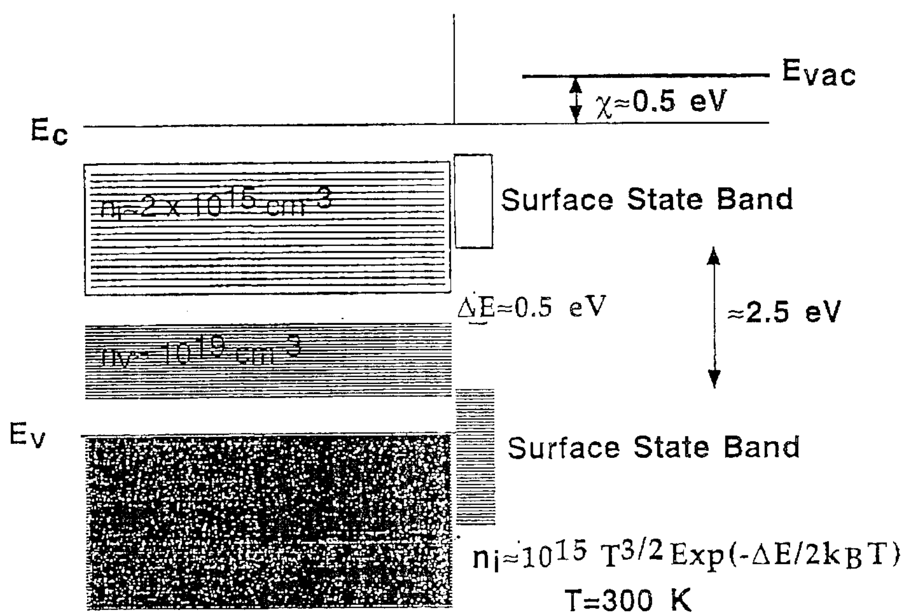

Fig. 10. Schematic diagram showing the proposed defect bands in the band gap of diamond. The locations of the surface state bands are also indicated.

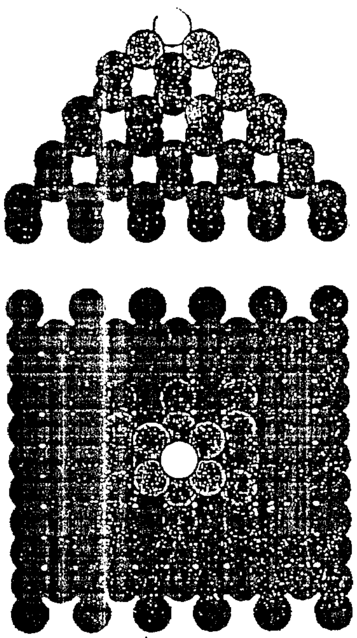

Fig. 12. The side (top panel) and top (bottom panel) view of the equilibrium atomic configuration of the Si cluster.

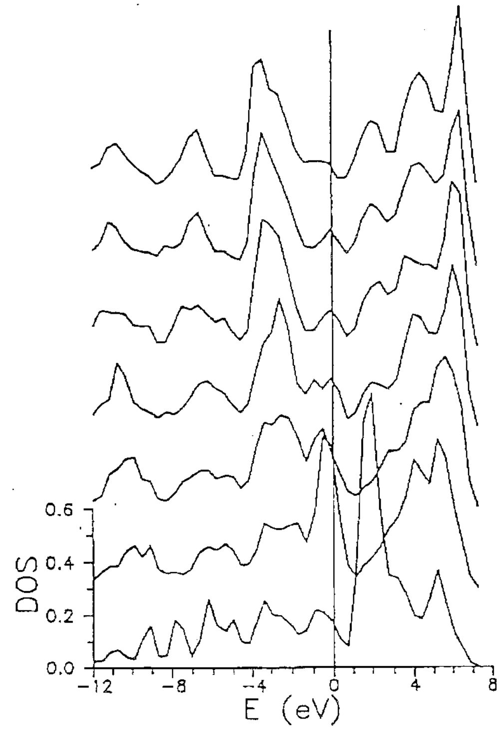

Fig. 13. Plot of the LDOS of atoms close to the symmetry axis. From the bottom to the top are the LDOS of the topmost, second..., and seventh layer atom. The zero on the energy scale corresponds to the highest filled state.

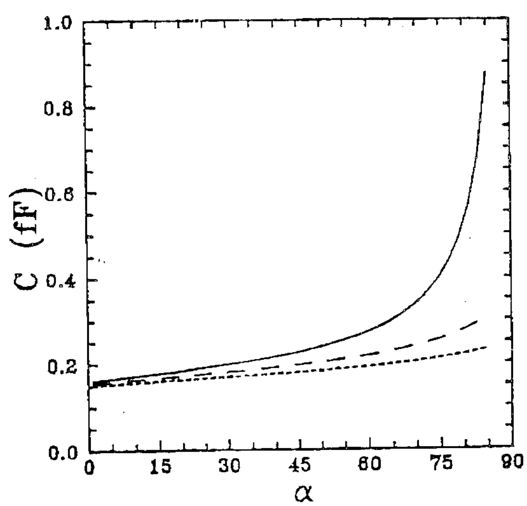

Fig. 14. Plot of C as a function of half-angle of the cone α, for R_1=2μm, R_2=0.05μm and Z_{cut}=-10 cosα. The solid, dash and dotted lines correspond to z=0, 0.5, and 1μm, respectively.

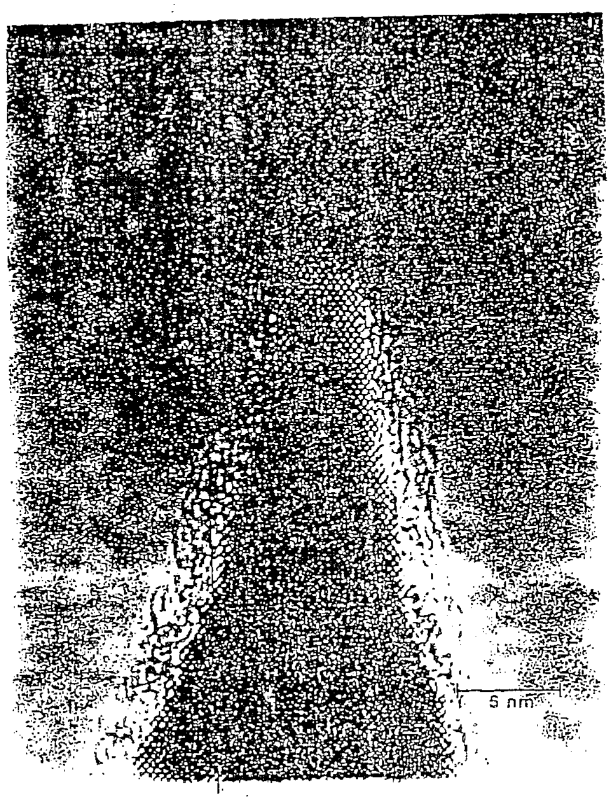

Fig. 15. An atomically sharp silicon emitter tip imaged by TEM formed using stress inhibited oxidation.

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Graduate Students

- Jun He, Ph.D. Thesis in Physics, "Theory of Electron Emission from Atomically Sharp Metallic Emitters in High Electric Fields," February, 1992, The Pennsylvania State University, Department of Physics, PhD, non-US

- Brock Weiss, M.S. Thesis in Physics, "Field Electron Energy Distributions for Atomically Sharp Metallic Emitters," June, 1994, The Pennsylvania State University, Department of Physics, MS, US

- R. Heiman, Temple University, Department of Electrical Engineering, MS, US

- J. D. Zuber, Temple University, Department of Electrical Engineering, PhD, US

- T. Cuff, Temple University, Department of Electrical Engineering, PhD, US

- T. H. Ting, Temple University, Department of Electrical Engineering, PhD, non-US

- Joseph Sherwin, THe Pennsylvania State University, Department of Physics, MS, US

Post-Doctoral Appointment

- Dr. Zhi-Hong Huang, The Pennsylvania State University, Department of Physics, non-US, 1993-1995

Short-Term Visitor

- Dr. Moon S. Chung, University of Ulsan, Ulsan, Korea, Department of Physics, non-US (no-cost participant), 1991-1995